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The Bioaccessibility of Lead (Pb) from Vacuumed House Dust on Carpets in Urban Residences

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Risk assessments for toxicants in environmental media via oral exposure often rely on measurements of total concentration in a collected sample. However, the human digestive system cannot dissolute all of a toxicant present in the binding matrix, and cannot absorb it with nearly 100% efficiency. *In vitro* bioaccessibility has been developed as a method to estimate oral bioavailability of a toxicant using a physiologically-based extraction procedure. Bioaccessibility measurements are more physiologically relevant than strong acid leaching measurements of concentration. A method for measuring bioaccessible lead in house dust was derived from the bioaccessibility method currently used for heavy metals in contaminated soils. House dust was collected from carpets in typical urban residences. Bioaccessible lead was measured in house dust (<75 μm) from the homes of 15 participants. The bioaccessibility ranged from 52.4% to 77.2% in gastric fluid, and 4.9% to 32.1% in intestinal fluid. House dust samples from five homes were analyzed to assess the relationship among lead bioaccessibility of three particle size fractions (<75, 75–150, and 150–250 μm). Changes in lead bioaccessibility as a function of particle size fraction were not significant for gastric fluid ($p = 0.7019$); however they were significant for intestinal fluid ($p = 0.0067$). This decrease of bioaccessibility may result from the readsorption of dissolved lead onto the dust particles or precipitation of lead with phosphates in a high-pH environment. The bioaccessibility data obtained for two biofluids were applied to the IEUBK model, and results for intestinal bioaccessibility of lead provide support for the model default value of 30% lead bioavailability of dust as a reasonable population indicator for dose, but the higher values for gastric bioaccessibility of lead appeared to provide an upper bound that approached actual blood lead levels in the children living in the studied homes. This upper bound seemed to overcome some of the limitations of the model when it lacks child-specific activity data and characterization of all exposure routes.

KEY WORDS: Bioaccessibility; bioavailability; house dust; lead

1. INTRODUCTION

House dust has been investigated to identify a proximate source of toxicants and quantify levels of toxicants in an exposure pathway that can be used for the estimation of human exposure in indoor residential settings.(1) Toxicants present in house dust include semivolatile and nonvolatile pesticides, PAHs (polyaromatic hydrocarbons), heavy metals, asbestos, and persistent organic compounds.(1) The specific information can be obtained from measurements of

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total level of each toxicant in house dust. The values are usually represented as dust loading ($\mu g/ft^2$, $\mu g/cm^2$) or dust concentration ($\mu g/g$) collected by wipe and vacuum sampling.[1] Since the ingestion of house dust can be a major route of children's nondietary exposure to lead,[2] data on the bioaccessible fraction of toxicants, like lead, present in house dust matrices can provide valuable new information on the exposure. It should also be a better indicator of the dose available to an individual or population (e.g., young children) from house dust.

House dust contaminated with lead is currently a primary source of childhood exposure to lead in the United States.[2–4] Hand-to-mouth activity is generally considered as the most important exposure pathway for young children.[5] It is well known that children are especially vulnerable to the effects caused by lead exposure because their rapidly developing nervous systems are particularly sensitive to lead.[6] Moreover, lead exposure to a fetus has been correlated with slow mental development and lower intelligence later in life.[6] Thus, exposure to lead can have a wide range of effects on a child's development and behavior.[7] For example, after exposure to small amounts of lead levels, children may appear inattentive, hyperactive, and irritable.[6] Children with high lead levels can have problems with learning and reading, and hearing loss.[6] At very high levels, lead can cause permanent brain damage and even death.[6]

A method for measuring bioaccessible lead in house dust used in this study was derived from a bioaccessibility method developed for heavy metals present in contaminated soils. A laboratory of Exposure Science Division in Environmental and Occupational Health Sciences Institute (EMAD-EOHSI) has successfully developed the bioaccessibility method of heavy metals (Pb, As, Cr, and Cd) and low-level radionuclides (^{137}Cs and ^{90}Sr) in various soil samples.[8–11] The method was modified for use with a different sample matrix (vacuumed house dust for *in vitro* extraction). The target for the current study was bioaccessible lead in house dust, and the levels were measured in both simulated gastric and intestinal fluid.

Two hypotheses were tested in the study. First, the levels of bioaccessible lead in vacuumed house dust (sieved to <75 μm) were different from lead levels obtained for total lead extracted from each sample, and the bioaccessible fractions were different between gastric and intestinal fluid extraction. Previous *in vitro* lead bioavailability studies for household dust,[12] soil,[13–16] and slagged aggregate[17] showed that lead dissolution was not 100%, and there was a difference in bioaccessible lead levels at different pH values. To quantify the bioaccessible lead in the artificial human stomach and small intestine, the bioaccessibility of lead was measured in simulated gastric and intestinal fluid.

The second hypothesis was that lead present in smaller dust particles had higher levels of bioaccessibility than lead associated with larger dust particles. Studies have shown that lead is generally concentrated in the fine fraction of dust,[18] and lead absorption within the body is inversely related to particle size.[18] Empirical evidence, however, does not necessarily indicate that collecting and analyzing the fine fraction of dust are a better predictor of children's blood lead level than total dust.[18] To link the bioaccessibility of lead with particle size fractions, the bioaccessible lead in two biofluids and total lead in house dust was measured in three particle size ranges: <75, 75–150, and 150–250 μm.

2. MATERIALS AND METHODS

2.1. Test House Dusts

Each of the 15 house dust samples was obtained from 50 homes that participated in the Evaluation of Carpet Dry Steam Cleaning (ECSC) project that was sponsored by U.S. HUD, Department of Housing and Urban Development. The ECSC study was designed to evaluate two different cleaning methods, HEPA vacuum only versus HEPA vacuum + dry steaming, for their effectiveness in removing lead and other toxicants from participant's carpets. Samples were taken before and after each home was cleaned, and then the samples were analyzed for lead and other materials in the house dust. Carpets were located in typical northern New Jersey residences during the period of July 2003 to June 2004. A participant had to have at least one child with an actual blood lead level higher than 15 μg/dL, which had been reported to the State Department of Health between 2000 and 2002. Dust samples were collected in dust mite allergen bags of a HEPA vacuum cleaner (HEPA Vacuum Cleaner Upright, Hoover®, Newton, IA) from the carpets. All sampled vacuum bags were shipped directly to EOHSI and stored in a cold room (constant temperature at 4°C and relative humidity at 35%) before analysis.

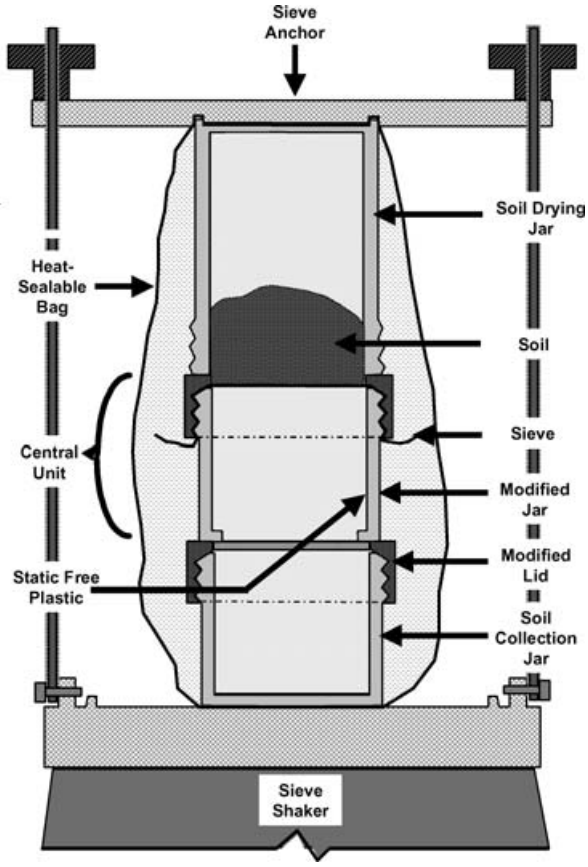

Fig. 1. Soil/house dust sieve unit (taken from Ellickson *et al.*, 2002).

2.2. House Dust Preparation

House dust vacuum samples collected from 15 residences were sieved to the <75-μm-diameter fraction using the unit shown in Fig. 1. This mass fraction was used as the primary sample for conducting the lead bioaccessibility experiments. The total lead and the lead that was present and bioaccessible in the gastric and intestinal fluids were measured in all samples. Five of these house dust samples were also sieved into three separate fractions (<75, 75–150, and 150–250 μm). Analyses were completed on each size fraction to determine which particle size fraction had the highest amount of bioavailable lead. The dust sieving was conducted using the standard operation procedures employed by the ESD-EOHSI bioaccessibility laboratory.[11]

2.3. Total Lead Dissolution for Lead Analysis

The total lead content in house dust was determined using the microwave-assisted digestion with atomic absorption analysis, and these data were used as the reference total lead concentration in each dust sample. Subsequently, the total lead concentration was used as the denominator in all calculations to determine the bioaccessible fraction of lead in house dust. Two subsamples of house dust (≈0.2g from each prepared dust sample) were transferred into 50 mL centrifuge tubes. Twenty milliliters of nitric acid (TraceMetal grade, Pb < 0.1 ppb, Fisher Scientific, Edison, NJ) and 10 mL of deionized water were added to each tube. Samples were digested in microwave oven (CEM Corporation, Matthews, NC) with the three-step program (1st stage of 90% power for 20 minutes, 2nd stage of 0% power for 20 minutes, and 3rd stage of 90% power for 20 minutes). After processing in the microwave oven, the samples were allowed to cool down in a hood, and then diluted up to 45 mL.

2.4. Bioaccessibility Extraction

The *in vitro* determination of bioaccessible lead was conducted using the modified version of the method[9–11] developed for measuring elemental bioaccessibility in soil.[13–15] The assay consisted of sequential extractions using synthetic human saliva, gastric, and intestinal fluid. Each step was completed at physiological temperature (around 37°C) and a mixing speed of 90 rpm. All bioaccessibility extraction experiments followed the cited standard operation procedures.[11]

2.4.1. Artificial Fluids Preparation

The artificial saliva used has been considered as the best correlate for natural saliva tests for extraction of gold and silver-tin amalgams in orthodonic appliances.[9] A modified version of this artificial saliva formula was developed by Hamel *et al.*[9] It was prepared using the following mixtures of 0.004M of calcium chloride dihydrate, 0.4% (w/v) of mucin, 0.005M of potassium chloride, 0.007M of sodium chloride, 0.004M of sodium phosphate dibasic, and 0.007M of urea. The artificial saliva was stored at 4°C prior to use.

The synthetic gastric fluid used by Hamel *et al.* (1999) was prepared following the recommendations of the U.S. Pharmacopoeia, which is employed in drug dissolution studies. The gastric fluid was prepared on the day of sample analysis using the following reagents: 0.03M of sodium chloride, 0.084M of hydrochloric acid, and 0.32% (w/v) of pepsin.

The synthetic intestinal fluid analogue was modified from the U.S. Pharmacopoeia formulation. This was done to provide a fluid with strong solvent properties. The artificial intestinal fluid used in these experiments was a 0.2M of solution of sodium bicarbonate.[9]

2.4.2. In Vitro *Extraction*

The amount of each house dust sample used in lead bioaccessibility experiment was based on soil/dust intake rate (0.135 g/day) for children between one and four years .[19] Lead dissolution in synthetic gastric juice was studied previously by Hamel *et al.* (1998), who showed that the lead solubility was relatively consistent within liquid-to-solid ratios of 100:1 to 5000:1 mL/g for simulated soil ingestion of 0.05 to 0.5 g. A 0.135 g of house dust sample was chosen as the amount of house dust ingested and 400:1 mL/g (50 + 4 mL/0.135 g) liquid-to-solid ratio was used throughout the extraction procedure.

Approximately 0.135 g sample of sieved house dust (triplicate samples for gastric and intestinal, respectively) was mixed with 4 mL of the artificial saliva. This was followed by the addition of 50 mL of gastric juice (pH = 1.4 ± 0.2 throughout the extraction). Then, the samples were placed into a constant temperature water bath (MSB-1122A-1, General Signal, Blue Island, IL) at 37°C and shaken for 2 hours at 90 rpm. After the saliva-gastric incubation, all samples were taken from the water bath. Subsequently, the gastric samples were filtered using a 0.45 μm of pore-sized cellulose nitrate filter in a polycarbonate 250 mL filter funnel apparatus. With the addition of 1 mL of nitric acid, the filtered aqueous gastric samples were preserved in polyethylene bottles. Meanwhile, the intestinal samples were adjusted to a pH 6.5 by adding 50 mL of intestinal juice. The samples were incubated again in the water bath at the same conditions. After the saliva-gastric + intestinal incubation, the intestinal samples were also filtered, and preserved in polyethylene bottles with 2 mL of nitric acid. Following the sequential gastrointestinal extraction, the house dust residue was captured on a filter. The residual was used to determine the total recovery and obtain a mass balance of lead for gastric and intestinal samples. The captured house dust residual samples followed the same procedures of total lead dissolution with 10 mL of nitric acid and 5 mL of deionized water, before the digestion of microwave oven. The lead bioaccessibility was calculated by dividing the lead concentration of each filtered fluid with total lead concentration found in each house dust sample. The schematic diagram (Fig. 2) shows the processes of *in vitro* extraction step by step.[11]

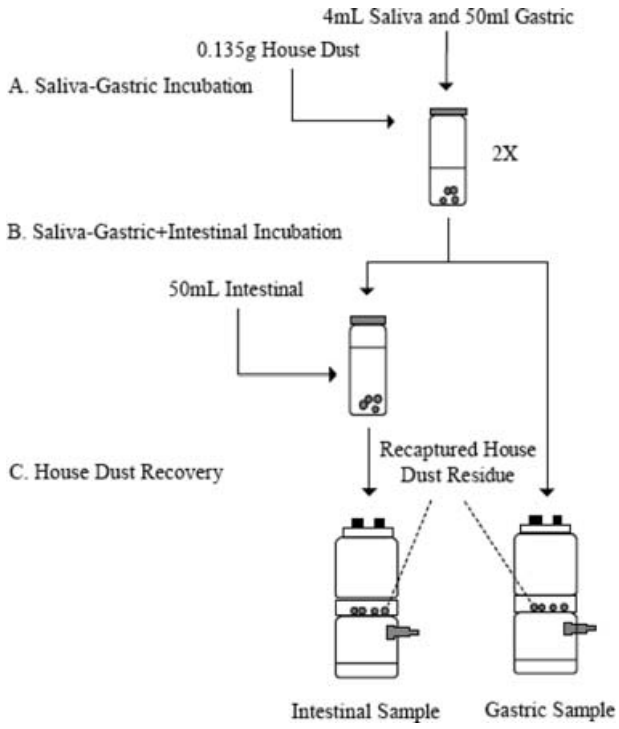

Fig. 2. Schematic representation of the bioaccessibility procedure (modified from Ellickson *et al.*, 2002).

2.5. Analytical Analysis

The gastric and intestinal extracts were directly transferred into centrifuge tubes from the polyethylene bottles, and diluted from 10 to 100 times prior to lead analysis. A 10-mL sample of both microwave-digested total lead and house dust residual sample was decanted, spun in a Metpath tabletop centrifuge (906 × g) for 10 minutes, and then diluted to be within the range of calibration curve from 2.5 to 25 ppb. A GFAA (Graphite Furnace Atomic Absorption Spectrometer; Perkin Elmer Zeeman 4100ZL, Norwalk, CT) with a detection limit of 2.5 ppb in solution was used to quantify the lead levels. The GFAA was calibrated for each run with standard prepared in Optima grade acid (Fisher Scientific). The National Institute of Standardized Testing (NIST; Gaithersburg, MD) reference material 1643D (Pb dissolved in water with a concentration of 18.15 ± 0.64 μg/L, density of the solution = 1.016 g/mL at 22°C) was used for quality control at each run of the GFAA analysis. Sample

Table I. Normality Test (Shapiro-Wilk Test with Total 45 Data)

Experiment	Tested Group	Test Statistic	*p*-Value
15 home samples	Gastric bioaccessibility	0.900696	0.0010
	Intestinal bioaccessibility	0.767012	<0.0001
5 home samples	Gastric bioaccessibility	0.965795	0.2025*
	Intestinal bioaccessibility	0.910170	0.0020

*Insufficient evidence to reject the assumption of normality.

digestion blanks and NIST 2710 soil spikes were included in all analytical runs. The allowable instrument error was ±20%, although most quality control results were within ±10%.

2.6. Statistical Analyses

The gastric and intestinal data ($N = 45$) were tested for normality using a Shapiro-Wilk test. The test for normality was conducted on 15 house dust samples sieved to <75 μm, and five house dust samples that were separated into particle size fractions of <75, 75–150, and 150–250 μm. The results presented in Table I show that three out of four experimental data sets were not normally distributed ($p < 0.05$), consequently further statistical analyses were done using nonparametric methods. All statistical analyses were provided by SAS® Program v9.1 (SAS Institute Inc., Cary, NC) and SigmaStat® Program v3.11 (Systat Software Inc., Richmond, CA).

2.7. IEUBK (Integrated Exposure Uptake Biokinetic Model for Lead in Children) Predictions

The U.S. EPA's IEUBK model estimates the potential concentration of lead in blood for a hypothetical child or population of children (six months to seven years old) based upon inputs of relevant variables, including absorption parameters, and rates of intake and exposure.[19] However, errors can occur due to assumptions made on lead bioavailability as well as other variables. The bioaccessible fractions of lead in gastric and intestinal fluids determined for house dust in this study were evaluated to determine the default input values. The values, used as determinants for GI/bioavailability, should be augmented in the model.

Two parts of the IEUBK were modified to use the house-specific bioaccessibility as input data. (1) The *GI Values/Bioavailability Information* option on the *Parameter Input* menu allowed the user to make adjustments to the gastrointestinal absorption coefficient to account for site-specific information on bioavailability.[19] (2) The *Blood Pb Versus Media Concentration* option on the *Computation* menu allowed the user to generate a plot of the relationship between blood lead concentration and the exposure to lead in a specific, user-selected, environmental medium (e.g., soil, dust, air, drinking water, or diet).[19]

Using the results from the lead bioaccessibility analyses, blood lead levels were predicted by substituting the default absorption value with the average bioaccessible lead values of 65% for gastric bioaccessibility and 12% for intestinal bioaccessibility. The input parameters for total lead levels in house dust were changed, ranging from 209 mg/kg as the lowest lead concentration of tested house dust to 1770 mg/kg as the highest concentration. Age of the child was selected to be between 0 and 84 months. The model simulation was run 15 times (maximum allowed by the program) to estimate the blood lead concentrations within a population of children exposed to bioaccessible lead levels found in house dust.

3. RESULTS AND DISCUSSION

3.1. Bioaccessibility Test for Vacuumed House Dusts

Fifteen house dust samples were chosen from the ECSC study and analyzed for bioaccessible lead (Table II). This sample size enables a statistical power of 80% with the significance level ($\alpha = 0.05$). The lead that was soluble in gastric fluid yielded the gastric bioaccessibility values that ranged from 52.4% (Home 027) to 77.2% (Home 009). After the solution passed into the intestinal phase of the simulated digestion, the bioaccessibility of the lead was reduced, and the values ranged from 4.9% (Home 013) to 32.1% (Home 010). The levels dropped abruptly below gastric bioaccessibility values since the pH increased to 6.5 with the addition of $NaHCO_3$ solution. Thus, part of the mobilized lead was removed from the intestinal solution. The difference between gastric bioaccessibility and intestinal bioaccessibility was statistically significant (Wilcoxon two-sample test; two-sided; $p < 0.0001$). Such a pH-dependent decrease in lead solubility (Fig. 3) has been observed by previous lead bioaccessibility tests on soil sets,[10,13–16] household dust samples,[12] and slagged aggregates.[17]

Table II. The Bioaccessibility Test for 15 Vacuumed House Dust Samples (Mean ± SD%)

Home ID	Total Lead Concentration (μg/g)	Gastric Bioaccessibility (%)	Intestinal Bioaccessibility (%)
001	1097	76.6 ± 3.1	6.7 ± 0.3
002	596	74.3 ± 14.5	6.7 ± 0.2
004	216	70.1 ± 4.5	8.5 ± 0.7
005	1125	68.9 ± 0.5	17.8 ± 0.7
006	358	71.3 ± 3.5	7.3 ± 1.1
007	1312	74.7 ± 2.2	6.0 ± 0.2
009	709	77.2 ± 0.9	11.1 ± 0.2
010	487	52.7 ± 1.4	32.1 ± 0.7
013	732	55.8 ± 1.4	4.9 ± 0.2
014	209	57.0 ± 1.9	9.3 ± 0.7
016	820	72.6 ± 2.7	5.75 ± 0.2
021	676	59.9 ± 2.0	12.3 ± 0.7
023	1770	55.1 ± 2.1	6.4 ± 0.2
027	813	52.4 ± 1.1	28.8 ± 2.1
029	541	53.7 ± 0.3	17.7 ± 0.9
Mean ± SD	764.1 ± 422.5	64.8 ± 9.4	12.1 ± 8.2

Simulated gastric juice resulted in higher bioaccessible lead than the intestinal samples (with the intestinal juice added) because of the strong acidity (pH = 1.4 ± 0.2 throughout the extraction) associated with the artificial gastric juice. The strong acid solution used in these experiments was representative of a fasting child to maximize lead dissolution. Thus, the results provided an upper-bound estimate of available lead that would apply to ingestion of small particles due to mouthing behavior by children several hours after food ingestion or under fasting conditions.[20]

Based on studies that determined the lead solubility in soils, the lead bioaccessibility appears to depend on chemistry (e.g., pH in the solution),[10,14–16,20] particle size distribution,[13] the mechanism of dissolution,[13] and the geochemistry of the soils.[17,21,22] The sharp decrease in lead bioaccessibility after passing to the simulated intestinal fluid would be due to one of three possibilities: (1) readsorption of lead onto the dust particles,[10,13,15] (2) complexation by pepsin,[10] or (3) chemical precipitation of the lead caused by phosphate within the higher pH environment found in the intestinal compartment.[10,13,15,20]

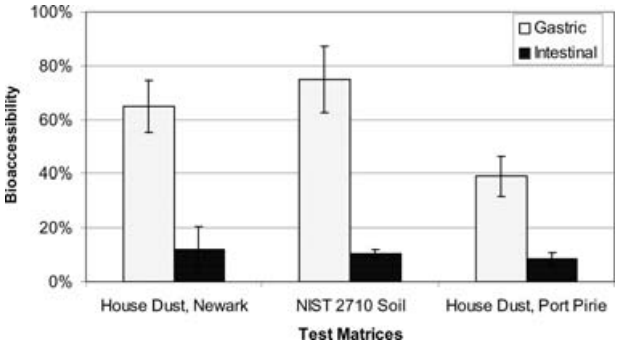

Fig. 3. The comparison of bioaccessible lead among three different test matrices. (a) House dust, Newark (<75 μm; N = 15) from current study results. (b) NIST 2710 Soil (<74 μm; N = 3) from Ellickson *et al.*, 2001. (c) House dust, Port Pirie (<53 μm; N = 7) in Australia study from Oliver *et al.*, 1999.

Since the lead bioaccessibility ranged from 52.4% to 77.2% for gastric extracts and 4.9% to 32.1% for intestinal extracts, the results suggest that the bioaccessibility of individual house dusts can be quite different. The current IEUBK risk assessment model for lead has adopted a 30% default value for the fraction of lead in house dust absorbed in the child's body, and the model does not consider the variation of house dust bioaccessibility. The range of house-specific bioaccessibility data found in these experiments can reduce the uncertainty in dose estimations for children by providing a range of values for sensitivity analyses, but supports the use of 30% lead absorption value as a determinant in the IEUBK model. The gastric bioaccessibility results, a much higher value of 65%, would provide a conservative (more protective) estimate of absorption. The use of the latter value may be reasonable in sensitivity analyses since the IEUBK did not predict any blood lead levels above 15 μg/dL using 30% lead absorption. Thus, the absence of site-specific data on activities and exposure routes may underestimate predicting a child's blood lead level. The value of 65% absorption may be useful in presenting simulations under prediction of blood lead values. However, these observations would suggest further evaluation and performance tests for use in the model.

3.2. Bioaccessibility/Recovery Test for Different Size Fractions

Lead in house dust is generally considered to be more concentrated in the fine fraction of dust, and lead absorption into the body is inversely related to particle size.[5] However, data showing a relationship between lead absorption and particle size are not readily available. The *in vitro* lead bioaccessibility was determined for three particle size fractions in 5 of the 15 house dust samples. The analyses were designed to examine changes of lead bioaccessibility in the three different particle size fractions.

Table III. The Bioaccessibility and Recovery (Mean ± SD%) of Vacuumed House Dust Samples for Three Subfractions (<75, 75–150, and 150–250 μm)

Home ID	Matrix (μm)	Gastric		Intestinal	
		Bioaccessibility (%)	Recovery (%)	Bioaccessibility (%)	Recovery (%)
001	<75	76.6 ± 3.1	85.1 ± 2.4	6.7 ± 0.3	71.2 ± 4.2
	75–150	71.0 ± 9.9	87.4 ± 4.0	15.6 ± 1.0	96.4 ± 13.2
	150–250	63.8 ± 7.1	91.4 ± 2.8	15.2 ± 3.7	94.6 ± 1.4
002	<75	74.3 ± 14.5	84.6 ± 13.3	6.7 ± 0.2	117.3 ± 12.6
	75–150	68.5 ± 4.2	83.3 ± 4.1	8.8 ± 0.6	115.9 ± 7.9
	150–250	71.2 ± 11.0	82.4 ± 8.6	4.7 ± 1.0	96.8 ± 10.6
004	<75	70.1 ± 4.5	76.5 ± 2.4	8.5 ± 0.7	76.6 ± 8.1
	75–150	69.3 ± 3.1	77.0 ± 1.4	9.9 ± 1.1	80.9 ± 3.4
	150–250	79.2 ± 21.0	86.1 ± 16.0	15.4 ± 5.8	76.7 ± 26.8
005	<75	68.9 ± 0.5	75.4 ± 4.3	17.8 ± 0.7	98.7 ± 3.7
	75–150	85.9 ± 7.2	87.7 ± 5.5	27.2 ± 4.9	121.8 ± 11.1
	150–250	90.9 ± 7.2	84.4 ± 4.6	26.8 ± 4.6	118.9 ± 1.8
006	<75	71.3 ± 3.5	78.2 ± 3.5	7.3 ± 1.1	96.8 ± 2.9
	75—150	56.3 ± 7.5	76.0 ± 5.7	23.9 ± 4.7	84.0 ± 5.1
	150–250	65.0 ± 6.0	79.9 ± 1.7	19.1 ± 6.9	93.5 ± 8.1

Recovery tests were also included as part of the bioaccessibility procedures for both artificial fluids and each of the three particle size fractions. The recovery tests established whether or not much lead was lost during the *in vitro* extraction. The bioaccessibility/recovery results are provided in Table III. The results for the recovery tests conducted for each fluid showed that the *in vitro* extraction procedures did not lose much lead in the sequential extraction system.

3.3. Bioaccessibility Results for the Three Size Fractions

The values determined for gastric bioaccessibility of three particle size fractions were not significantly different (Kruskal-Wallis test; $p = 0.7019$). However, the intestinal bioaccessibility obtained for each of the three particle size fractions was significantly different (Kruskal-Wallis test; $p = 0.0067$). Further, a nonparametric post hoc multiple comparison test (Tukey's method for balanced subgroups; $\alpha = 0.05$) was completed to compare the intestinal bioaccessibility obtained for each size fraction, and there were statistical differences among the size fractions (Table IV).

The intestinal bioaccessibility of particle size fraction sieved to <75 μm was significantly lower ($p < 0.05$) than the values obtained for both 75–150 μm and 150–250 μm particle size fractions; however, the difference was not significant between the size fraction of 75–150 μm and 150–250 μm. The gastric bioaccessibility results obtained for each size fraction suggested that lead dissolution was not affected by the dust particle size; however, lead dissolution for each size fraction was influenced by intestinal fluid. This behavior seems to indicate that lead is dissolved uniformly in gastric fluid but can be quickly readsorbed onto the surface of smaller dust particles after the addition of artificial intestinal juice. The magnitude of

Table IV. Nonparametric Tests for Lead Bioaccessibility by Particle Size Fractions (Kruskal-Wallis ANOVA Test & Tukey's Multiple Comparison Test)

	Matrix (μm)	No.	Mean Score	Pr < χ^2 *p*-Value	Min.	Med.	Max.	Grouping
Gastric	<75	15	24.600	(0.7019)	0.577	0.719	0.848	
	75–150	15	20.733		0.490	0.687	0.941	
	150–250	15	23.667		0.561	0.689	1.001	
Intestinal	<75	15	14.333	(0.0067)	0.062	0.072	0.183	A
	75–150	15	28.467		0.081	0.157	0.319	B
	150–250	15	26.200		0.038	0.141	0.320	B

Note: Different letters indicate significant differences ($p < 0.05$) by balanced Tukey's multiple comparison test; the same letter indicates no difference between median ranks.

adsorption of dissolved lead onto the dust particles is inversely related to particle size, because smaller particles have larger surface area at unit mass.[5,23] Intestinal recovery data also supported the concept that the larger difference between gastric and intestinal bioaccessibility resulted from the absorption of dissolved lead onto dust particles or chemical precipitation in the mixture caused by phosphates in a high pH environment.[10,13,15,20]

3.4. IEUBK Model Estimates of Dose Using Bioaccessible Lead

The IEUBK model predicted two levels of blood lead concentrations: one using gastric bioaccessibility and the other using intestinal bioaccessibility (Fig. 4). It is believed that most lead is absorbed into the upper part of the intestine,[6] consequently the values for intestinal bioaccessibility should be more representative of the absorption fraction. However, the gastric bioaccessibility provides a maximum fraction of lead available within the human gastrointestinal system. The IEUBK model predictions showed that blood lead levels generally would not exceed the EPA/CDC level of 10 μg/dL using mean value of intestinal bioaccessibility (12%) or default value (30%) in the range of tested house dust concentrations. However, the calculated values based on mean value of gastric bioaccessibility (65%) easily surpassed the limit of 10 μg/dL.

The IEUBK predictions, based on model default parameter of 30% GI absorption, were inconsistent with the reported blood lead levels found among the children in these homes. The model predictions using the 30% absorption were expected to approach the blood lead levels (>15 μg/dL) reported for each child; but conversely, the predictions could not reach these levels. However, the 65% absorption value associated with the gastric bioaccessibility did approach the actual blood lead level values that ranged from 11 to 32 μg/dL. The inconsistency between the actual blood lead levels and the model predictions could be due to many factors. First, it may be derived from the difference between the time for each child's blood lead level screening test measurements (at least six months ahead of sampling schedule) and the time at which lead concentration was determined in the house dust from carpets. Second, a child might be exposed to lead present in residences, but some route(s) could not be quantified for these analyses. Third, the home environment might be improved with the vacuum cleaning of carpets or abatement of leaded paint inside the house after the child's screening test result was reported to the State Department of Health.

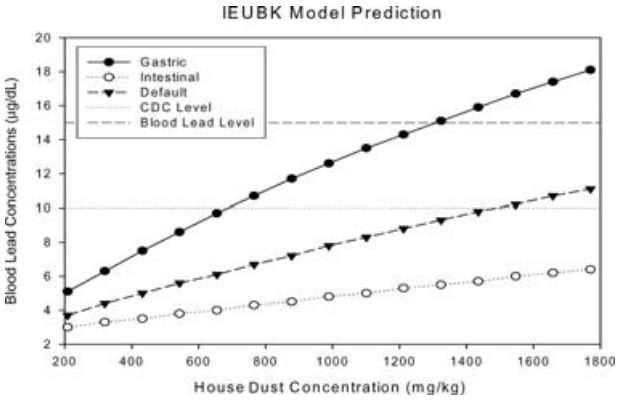

Fig. 4. The comparison of IEUBK model predictions for obtained gastric bioaccessibility (65%), intestinal bioaccessibility (12%), and model default value (30%).

The default value for GI/bioavailability (30%) in the IEUBK model is considered a reasonable estimator for predicting the blood lead levels in a target population. However, the results from this study suggest that sensitivity analyses should include the range of absorption values <15% to >60%. This will provide a better indicator on possible lead levels in the blood. The highest value of intestinal bioaccessibility (32%) is essentially equivalent to the default absorption value used by the IEUBK model. A one-sample median test was conducted to determine whether or not a sample median differs significantly from a hypothesized value. The IEUBK model default parameter, 30% lead absorption from soil/dust, was assumed as a hypothesized value; it was significantly different with the obtained intestinal bioaccessibility data (Wilcoxon signed rank test; $H_0 = 0.3$; $p < 0.0001$). The percentiles and confidence intervals (95%) corresponding to hypothesized 30% intestinal bioaccessibility were obtained and reported in Table V. The IEUBK model default value covers over 90% of all intestinal bioaccessibility values determined in our experiments; however, the 30% did not predict the actual blood lead levels above 15 μg/dL, which was measured in each child. Thus, although the default value used for IEUBK risk estimates is a reasonable estimator of absorption for determining the blood lead levels in target populations, it may miss high exposure individuals because of the lack of other information on the specific child (activities) or routes of exposure.

Table V. The Percentile Corresponding to 30% Intestinal Bioaccessibility and Confidence Intervals (95%) for the Percentile

		95% Confidence Limits Distribution Free		Order Statistics		
Quantile	Estimate	Lower	Upper	LCL Rank	UCL Rank	Coverage
100%	0.3270					
99%	0.3270	.	.	.	.	.
95%	0.3128	0.2939	0.3270	41	45	82.76
90%	**0.2939**	**0.1801**	**0.3270**	**37**	**45**	**95.93**
75%	0.1663	0.1103	0.2939	29	41	95.46
50%	0.0841	0.0660	0.1129	16	30	96.43
25%	0.0631	0.0568	0.0668	5	17	95.46
10%	0.0568	0.0463	0.0616	1	9	95.93
5%	0.0504	0.0463	0.0568	1	5	82.76
1%	0.0463	.	.	.	.	.
0%	0.0463					

4. CONCLUSIONS

Through the successful modification of a soil bioaccessibility method to quantify the bioaccessible fraction of lead in environmentally contaminated house dust, it was found that the gastric bioaccessibility was significantly ($p < 0.0001$) higher than the intestinal bioaccessibility for house dust (<75-μm-diameter size). This was due to the readsorption of dissolved lead onto the surface of dust particles at high pH. In addition, the level of intestinal bioaccessibility was significantly different ($p = 0.0067$) among three house dust particle size fractions (<75, 750–150, and 150–250 μm), but the gastric bioaccessibility was the same for each particle size fraction ($p = 0.7019$). The lead bioaccessibility data obtained for two biofluids (gastric and intestinal fluids) can be applied to the IEUBK model, which provided a support for the model default value of 30% as a reasonable estimate for bioavailable lead in house dust. However, since the model did not predict the actual high blood lead values in the children, the 30% value should be accompanied by sensitivity analyses with a range of absorption from <15% to >60% to reduce underestimation of blood lead levels because of the lack of site-specific information on an individual's activities or all major routes of exposure.

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REFERENCES

1. Lioy, P. J., Freeman, N. C. G., & Millette, J. R. (2002). Dust: A metric for use in residential and building exposure assessment and source characterization. *Environmental Health Perspectives*, *110*(10), 969–983.
2. Lanphear, B. P., Weitzman, M., Winter, N. L., Eberly, S., Yakir, B., Tanner, M., Emond, M., & Matte, T. D. (1996). Lead-contaminated house dust and urban children's blood lead levels. *American Journal of Public Health*, *86*(10), 1416–1421.
3. Thornton, I., Davies, D. J., Watt, J. M., & Quinn, M. J. (1990). Lead exposure in young children from dust and soil in the United Kingdom. *Environmental Health Perspectives*, *89*, 55–60.
4. Rhoads, G. G., Ettinger, A. S., Weisel, C. P., Buckley, T. J., Goldman, K. D., Adgate, J. L., & Lioy, P. J. (1999). The effect of dust lead control on blood lead in toddlers: A randomized trial. *Pediatrics*, *103*, 551–555.
5. U.S. Department of Housing and Urban Development. (1995). *Guidelines for the Evaluation and Control of Lead-Based Paint Hazard in Housing*. HUD-1539-LBP. Washington, DC: U.S. HUD.
6. U.S. Department of Health and Human Services, Agency for Toxic Substance and Disease Registry. (1999). *Toxicological Profile for Lead*. Atlanta, GA: U.S. DHHS-ATSDR.
7. U.S. Environmental Protection Agency. (2001). *Protect Your Family from Lead in Your Home*. EPA747-K-99-001. Washington, DC: U.S. EPA.
8. Hamel, S. C., Buckley, B., & Lioy, P. J. (1998). Bioaccessibility of metals in soils for different liquid to solid ratios in synthetic gastric fluid. *Environmental Science & Technology*, *32*(3), 358–362.
9. Hamel, S. C., Ellickson, K. M., & Lioy, P. J. (1999). The estimation of the bioaccessibility of heavy metals in soils using artificial biofluids by two novel methods: Mass-balance and soil recapture. *Science of the Total Environment*, *243/244*, 273–283.
10. Ellickson, K. M., Meeker, R. J., Gallo, M. A., Buckley, B. T., & Lioy, P. J. (2001). Oral bioavailability of lead and

arsenic from a NIST standard reference soil material. *Archives of Environmental Contamination and Toxicology*, *40*, 128–135.
11. Ellickson, K. M., Schopfer, C. J., & Lioy, P. J. (2002). The bioaccessibility of low-level radionuclides from two Savannah River site soils. *Health Physics*, *83*(4), 476–484.
12. Oliver, D. P., McLaughlin, M. J., Naidu, R., Smith, L. H., Maynard, E. J., & Calder, I. C. (1999). Measuring Pb bioavailability from household dusts using an *in vitro* model. *Environmental Science & Technology*, *33*(24), 4434–4439.
13. Ruby, M. V., Davis, A., Kempton, J. H., Drexler, J. W., & Bergstrom, P. D. (1992). Lead bioavailability: Dissolution kinetics under simulated gastric conditions. *Environmental Science & Technology*, *26*(6), 1242–1248.
14. Ruby, M. V., Davis, A., Link, T. E., Schoof, R., Chaney, R. L., Freeman, G. B., & Bergstrom, P. D. (1993). Development of an *in vitro* screening test to evaluate the *in vivo* bioaccessibility of ingested mine-waste lead. *Environmental Science & Technology*, *27*(13), 2870–2877.
15. Ruby, M. V., Davis, A., Schoof, R., Eberle, S., & Sellstone, C. M. (1996). Estimation of lead and arsenic bioavailability using a physiologically based extraction test. *Environmental Science & Technology*, *30*(2), 422–430.
16. Oomen, A. G., Hack, A., Minekus, M., Zeijdner, E., Cornelis, C., Schoeters, G., Verstraete, W., Wiele, T. V. D., Wragg, J., Rompelberg, C. J. M., Sips, A. J. A. M., & Wijnen, J. H. V. (2002). Comparison of five *in vitro* digestion models to study the bioaccessibility of soil contaminants. *Environmental Science & Technology*, *36*(15), 3326–3334.
17. Davis, A., Ruby, M. V., Goad, P., Eberle, S., & Chryssoulis, S. (1997). Mass balance on surface-bound mineralogic, and total lead concentrations as related to industrial aggregate bioaccessibility. *Environmental Science & Technology*, *31*(1), 37–44.
18. U.S. Environmental Protection Agency. (1995). *Sampling House Dust for Lead—Basic Concepts and Literature Review*. EPA747R-95-007. Washington, DC: U.S. EPA.
19. U.S. Environmental Protection Agency. (2002). *User's Guide for the Integrated Exposure Uptake Biokintetic Model for Lead in Children (IEUBK) Windows® Version*. EPA 540-K-01-005. Washington, DC: U.S. EPA.
20. Oomen, A. G., Rompelberg, C. J. M., Bruil, M. A., Dobbe, C. J. G., Pereboom, D. P. K. H., & Sips, A. J. A. M. (2003). Development of an *in vitro* digestion model for estimating the bioaccessibility of soil contaminants. *Archives of Environmental Contamination and Toxicology*, *44*, 281–287.
21. Davis, A., Drexler, J. W., Ruby, M. V., & Nicholson, A. (1993). Micromineralogy of mine wastes in relation to lead bioavailability, Butte, Montana. *Environmental Science & Technology*, *27*(7), 1415–1425.
22. Ruby, M. V., Schoof, R., Brattin, W., Goldade, M., Post, G., Harnois, M., Mosby, D. E., Casteel, S. W., Berti, W., Caepenter, M., Edwards, D., Cragin, D., & Chappell, W. (1999). Advances in evaluating the oral bioavailability of inorganics in soil for use in human health risk assessment. *Environmental Science & Technology*, *33*(21), 3697–3705.
23. Lewis, R. G., Fortune, C. R., Willis, R. D., Camann, D. E., & Antley, J. T. (1999). Distribution of pesticides and polycyclic aromatic hydrocarbons in house dust as a function of particle size. *Environmental Health Perspectives*, *107*(9), 721–726.